

Quantum theory of Kerr nonlinearity with Rydberg slow light polaritons

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We study the propagation of Rydberg slow light polaritons through an atomic medium for intermediate interactions. Then, the dispersion relation for the polaritons is well described by the slow light velocity alone, which allows for an analytical solution for arbitrary shape of the atomic cloud. We demonstrate the connection of Rydberg polaritons to the behavior of a conventional Kerr nonlinearity for weak interactions, and determine the leading quantum corrections for increasing interactions. We propose an experimental setup which allows one to measure the effective two-body interaction potential between slow light polaritons as well as higher body interactions.

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Photons interact with its environment much weaker than other quanta and therefore represent an excellent carrier of information. On the other hand, a long-standing goal is the realization of a strong and controllable interactions on the level of individual photons. Such an interaction would pave the way towards ultralow-power all-optical signal processing [1, 2], which in turn has important applications in quantum information processing and communication [3–6]. A natural mechanism for an interaction is provided by the Kerr nonlinearity of conventional materials [7], but unfortunately is restricted to high intensities of the fields [8]. On the other hand, the appearance of a strong interaction between individual photons has been experimentally realized using Rydberg slow light polaritons. Here, we provide the theoretical framework to connect this regime of strong interaction with the phenomena of a classical Kerr nonlinearity.

Rydberg slow light polaritons have emerged as a highly promising candidate to engineer strong interactions between optical photons with a tremendous recent experimental success. A variety of applications were shown such as a deterministic single photon source [9], an atom-photon entanglement generation [10], as well as a single photon switch [11] and transistors [12–14]. Moreover, the regime of strong interaction between photons has been experimentally demonstrated leading to a medium transparent only to single photons [15], as well as the appearance of bound states for photons [16]. From theoretical point of view, the effective low energy theory is well understood from a microscopic approach [17, 18], but a full description of the propagation of photons through the medium is limited to extensive numerical simulations and low photon number [15, 16, 19–23].

In this letter, we provide the full input-output formalism of Rydberg polaritons for intermediate interaction strength but arbitrary incoming photon number and shape of the atomic medium. The analysis is performed in the regime with large detuning from the intermediate p-level, where losses are strongly suppressed and the effective low-energy theory for the polaritons is well de-

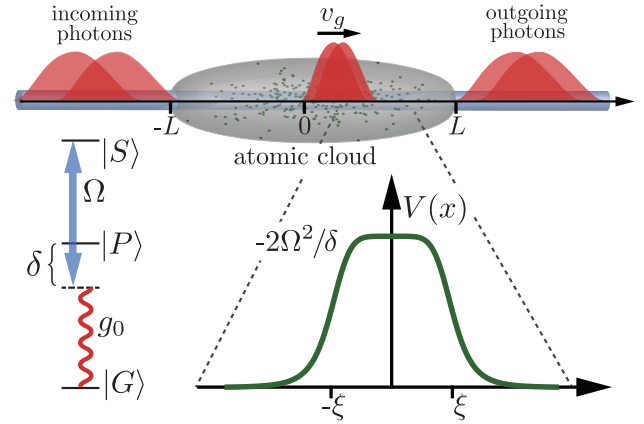


FIG. 1. Setup of Rydberg slow light polaritons: each atom consists of three relevant levels, ground state $|G\rangle$, intermediate p-level $|P\rangle$ and Rydberg state $|S\rangle$; the latter are coupled by a strong laser. Incoming photons with a single transverse channel enter the medium and are converted into slow light $v_g < c$ Rydberg polaritons. The interaction between the Rydberg states provides an effective interaction $V(x)$ for the polaritons.

scribed by an effective interaction potential [18]. We demonstrate the connection of Rydberg polaritons to the behavior of a conventional Kerr nonlinearity for weak interactions, and determine the leading quantum corrections for such a Kerr nonlinearity. We demonstrate the potential to experimentally determine the effective interaction potential as well as higher body interactions between the slow light polaritons within a homodyne setup.

It is important to point out that previous approaches to describe the quantum propagation of photons in a nonlinear Kerr medium based on a quantization of the phenomenological nonlinear equations provide an inconsistent quantum field theory [24–26]. This inconsistency was removed by requiring a non-local response in time, which in addition provides a noise term [27, 28]. For Rydberg slow light polaritons such a non-local response in time is absent, but the microscopic analysis naturally

provides a mass term accounting for deviation from the slow light velocity, as well as a finite range of the effective interaction potential describing the blockade phenomena. Both terms alone are sufficient to render the quantum theory well defined. As a consequence, we conclude that the proposed inability to generate a photonic phase gate by a large Kerr nonlinearity [28] does not apply to Rydberg slow light polaritons.

We consider a system of Rydberg slow light polaritons in the dispersive limit with large detuning $\delta > \gamma, \Omega$ from the intermediate p-level, see Fig. 1. Here, γ describes the decay rate of the p-level, while Ω denotes the Rabi frequency of the coupling laser. Within this regime, losses are strongly suppressed and the intermediate p-level can be adiabatically eliminated [18]. We are interested in the propagation of photons along a one-dimensional mode through the medium with frequency close to the condition of electromagnetic induced transparency. In the regime with a low density of Rydberg polaritons, the system is well described by an effective low energy quantum theory [18]. The interaction potential between the polaritons is characterized by a blockade radius ξ and the potential depth $2\hbar\Omega^2/\delta$ at short distances. For a microscopic van der Waals interaction with $C_6\delta < 0$ the effective interaction potential reduces to $V(x) = -(2\hbar\Omega^2/\delta)[1 + (x/\xi)^6]^{-1}$ with the blockade radius $\xi = (|C_6\delta|/2\Omega^2)^{1/6}$, see Fig. 1. Note, that for increasing polariton densities additional many-body interactions are expected to appear [29]. In the following, we mainly focus on the two body interactions, but the extension to include many-body interactions is straightforward and its influence is discussed in the last part.

The kinetic energy for the polaritons at low energies is determined by the slow light velocity of the polaritons and an effective mass term accounting for the curvature in the dispersion relation. The important aspect for the present analysis is the possibility to drop the mass term for moderate interactions between polaritons. The precise condition for the validity of this approximation is discussed below. Then, the Hamiltonian describing the propagation of photons through the spatially inhomogeneous medium with atomic density $n_a(x)$ is given by

$$H = \int dx [\beta(x)\psi^\dagger(x)] (-i\hbar c \partial_x) [\beta(x)\psi(x)] + \frac{1}{2} \int dx dy n(x)n(y)V(x-y)\psi^\dagger(x)\psi^\dagger(y)\psi(y)\psi(x). \quad (1)$$

Here, ψ and ψ^\dagger denote the bosonic field operators for the Rydberg slow light polaritons with $[\psi(x), \psi^\dagger(x')] = \delta(x-x')$. Furthermore, $\beta(x)$ describes the amplitude of the polariton to be in a photonic state and is related to the slow light velocity $v_g = c\beta(x)^2$, while $n(x) = 1 - \beta(x)^2$ is the probability for the polariton to be in the Rydberg state. These quantities are determined by the atomic density $n_a(x)$ via $\beta(x) = \Omega/\sqrt{\Omega^2 + g_0^2 n_a(x)}$ with g_0 the single

atom coupling. Note that outside the atomic medium the operator ψ describes non-interacting photons.

In the following, it is convenient to introduce a coordinate transformation which removes the reduced velocity v_g of the polaritons inside the media, i.e., we measure distances in the time z/c which is required for the polaritons to reach the position x . The coordinate transformation takes the form $z = \zeta^{-1}(x) = \int_0^x dy (1/\beta(y)^2)$, and the Hamiltonian reduces to

$$H = -i\hbar c \int dz \hat{\psi}^\dagger(z) \partial_z \hat{\psi}(z) + \frac{1}{2} \int dz dw \tilde{n}(z) \tilde{n}(w) \tilde{V}(z, w) \hat{\psi}^\dagger(z) \hat{\psi}^\dagger(w) \hat{\psi}(w) \hat{\psi}(z) \quad (2)$$

with $\tilde{n}(z) = n(\zeta(z))$, $\tilde{V}(z, w) = V(\zeta(z) - \zeta(w))$, and $\hat{\psi}^\dagger(z) = \psi^\dagger(\zeta(z))\beta(\zeta(z))$; the new operators $\hat{\psi}$ still satisfy the bosonic canonical commutation relations.

The quantum many-body theory in Eq. (2) is exactly solvable. This remarkable property is most conveniently observed by analyzing the Heisenberg equations for the field operator $\hat{\psi}(z, t)$,

$$i\hbar \partial_t \hat{\psi}(z, t) = -i\hbar c \partial_z \hat{\psi}(z, t) + K(z, t) \hat{\psi}(z, t) \quad (3)$$

with the operator $K(z, t)$ accounting for the interaction,

$$K(z, t) = \int dw \tilde{n}(z) \tilde{n}(w) \tilde{V}(z, w) \hat{\psi}^\dagger(w, t) \hat{\psi}(w, t). \quad (4)$$

In the following, we denote by $\hat{\psi}_0(z)$ the non-interacting field operator at time $t = 0$. Then, the interacting field operator $\hat{\psi}(z, t)$, satisfying the Heisenberg equation above, reduces to $\hat{\psi}(z, t) = e^{-i\hat{J}(z, t)} \hat{\psi}_0(z - ct)$ with the operator

$$\hat{J}(z, t) = \frac{1}{c\hbar} \int_{z-ct}^z dw \int_{-\infty}^{\infty} du \tilde{n}(w) \tilde{n}(u) \tilde{V}(u, w) \hat{I}(z - w + u - ct) \quad (5)$$

and the polariton density operator $\hat{I}(z) = \hat{\psi}_0^\dagger(z) \hat{\psi}_0(z)$.

We start by analyzing the behavior of the *two-photon* solution. It allows us to determine the influence of the involved approximations and to provide a connection to previous results on two-polariton propagation [16, 18]. For an arbitrary two photon state $|\phi\rangle$, the incoming wave function is defined via

$$\phi^{\text{in}}(x - ct, y - ct) = \lim_{t \rightarrow -\infty} \langle 0 | \hat{\psi}(z, t) \hat{\psi}(w, t) | \phi \rangle / \sqrt{2}, \quad (6)$$

with the coordinates $x = \zeta(z)$ and $y = \zeta(w)$, and the outgoing wave function ϕ^{out} via an analogous expression in the limit $t \rightarrow \infty$. Using the above exact solution for the bosonic field operators $\hat{\psi}(z, t)$, we obtain the relation between the incoming and the outgoing photon wave function

$$\phi^{\text{out}}(x, y, t) = e^{-i\varphi(x-y)} \phi^{\text{in}}(x - ct', y - ct') \quad (7)$$

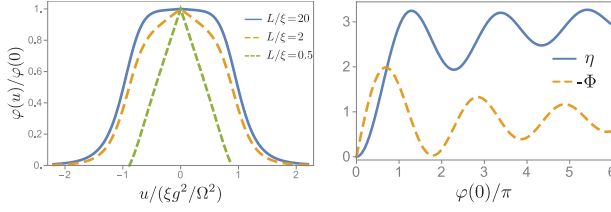


FIG. 2. (left) Phase factor $\varphi(u)$ for homogeneous distributions of atoms with different lengths L . For short clouds the condition $\xi \ll L$ is not satisfied and the behavior of $\varphi(u)/\varphi(0)$ is no longer universal, like it is for a long medium. (right) Phase shift and suppression of the electric field for coherent state in the limit $\xi_{\text{out}} \ll l_{\text{coh}}$ as a function of a single photon nonlinearity strength parametrized by $\varphi(0)$. For weak nonlinearities $\varphi(0) \ll 1$ the electric field suppression scales quadratically and the phase shift linearly with the nonlinearity strength. The suppression as well as phase shift oscillate with the increasing strength of interaction.

where $t' = t - \Delta t$ accounts for the delay of the polaritons inside the medium with $\Delta t = \int_{-\infty}^{\infty} dy (1/\beta(y)^2 - 1)/c$. Note, that the outgoing wave function only depends on the reduced coordinate $\tau_1 = x - ct'$ and $\tau_2 = y - ct'$; therefore, in the following, we will use these reduced coordinates to express the outgoing wave function. The phase factor $\varphi(u)$ describes the correlations built up between the photons during the propagation through the medium and takes the form

$$\varphi(u) = \frac{1}{\hbar c} \int_{-\infty}^{\infty} dw \tilde{n}(w+u) \tilde{n}(w) \tilde{V}(w+u, w). \quad (8)$$

It is instructive to analyze this phase factor for a specific homogeneous atomic density distribution $n_a(x) = \bar{n}_a \theta(L^2/4 - x^2)$ where θ is a Heaviside step function. The time delay simplifies to $\Delta t = L(1/\tilde{v}_g - 1/c)$, where the slow light velocity $\tilde{v}_g = c\Omega^2/(g^2 + \Omega^2)$ and the collective coupling between photon and matter $g = g_0\sqrt{\bar{n}_a}$. In turn, the phase shift acquires the peak value

$$\varphi(0) = \frac{g^4 V(0)L}{(g^2 + \Omega^2)\Omega^2 \hbar c} = -\frac{g^2}{g^2 + \Omega^2} \frac{\kappa\gamma}{\delta} \quad (9)$$

with κ the optical depth of the medium. The width of the signal in the phase $\varphi(u)$ is enhanced from the blockade radius by the slow light velocity to $\xi_{\text{out}} = \xi(g^2 + \Omega^2)/\Omega^2$. The exact phase shapes for different medium lengths are shown in Fig. 2. The determination of $\varphi(u)$ for other physical distributions of atoms is straightforward.

Note, that the interaction provides a spatially dependent phase factor correlating the photons, but is unable to induce a modification in the intensity correlations. A bunching of photons as observed in the experiments by Firstenberg et al. [16] requires the inclusion of the mass term. Here, we estimate the influence of this term, and determine the regime of validity for our approximation to drop it, for details see the supplement material. First, the inclusion of the mass would lead to an additional phase

shift estimated by $\varphi_m \sim \hbar\Delta t/(m\xi^2)|\varphi(0)^2 + i\varphi(0)| = |\varphi(0) + i|g^6/(g^2 + \Omega^2)^3 L^2/\xi^2|$, where we used the expression for the polariton mass $m = \hbar \frac{(g^2 + \Omega^2)^3}{2c^2 g^2 \Delta \Omega^2}$ [18]. In order to drop phenomena like the bunching of photons we require $\varphi_m \ll 1$. Secondly, we would like the phase shift induced by the interaction to dominate the behavior, i.e., $\varphi(0) \gg \varphi_m$. The two conditions are either satisfied for a weak coupling between photon and matter or for a short medium.

The two-photon analysis can be generalized straightforwardly to an N -photon Fock state. Then, the wave function reduces to

$$\frac{\phi^{\text{out}}(\tau_1, \dots, \tau_N)}{\phi^{\text{in}}(\tau_1, \dots, \tau_N)} = \exp \left[-i \sum_{i < j}^N \varphi(\tau_i - \tau_j) \right],$$

where $\tau_i = x_i - c[t - \Delta t]$. This allows one to derive the outgoing wave function for an arbitrary incoming state. Of special experimental interest however is the behavior of coherent states. A general incoming coherent state is characterized by its incoming electric field expectation value $\mathcal{E}(x - ct) = \lim_{t \rightarrow -\infty} E(x, t)$ with $E(x, t) = \langle \mathcal{E} | \psi(x, t) \beta(x) | \mathcal{E} \rangle$. Then, the outgoing electric field behaves as

$$\frac{E^{\text{out}}(\tau)}{\mathcal{E}(\tau)} = \exp \left(\int du |\mathcal{E}(u)|^2 \left[e^{-i\varphi(u-\tau)} - 1 \right] \right), \quad (10)$$

where $\tau = x - c[t - \Delta t]$. In the limit of a weak nonlinearity $\varphi(u) \ll 1$, we can recover the result of a classical Kerr nonlinearity. In this regime, the incoming wave packet has a size l_{coh} much larger than the characteristic size of the interaction $l_{\text{coh}} \gg \xi_{\text{out}}$, and propagates through a long medium $L \gg \xi$. Then, the Eq. (10) reduces to $E^{\text{out}}(\tau) = \mathcal{E}(\tau) \exp(-i\sigma |\mathcal{E}(\tau)|^2)$ with σ the strength of the Kerr nonlinearity. The latter depends on the shape of the atomic density distribution and reduces for a homogeneous atomic density to

$$\sigma = \int du \varphi(u) = \frac{2\pi}{3} \frac{g^2}{\Omega^2 + g^2} \frac{\kappa\gamma}{\delta} \xi_{\text{out}}. \quad (11)$$

However, it is important to stress, that Eq. (10) includes also the corrections to the Kerr nonlinearity due to the quantum fluctuations. The corrections can be analyzed by the full evaluation of the factor $i\Phi + \eta = -\int du (\exp(-i\varphi(u)) - 1)/\xi_{\text{out}}$, where Φ describes the strength of the Kerr nonlinearity, whereas η accounts for a suppression of the coherences due to quantum fluctuations, see Fig. 2. The latter follows from the fact that a coherent state is a superposition of different number states, where each number state picks up a slightly different phase factor.

The full characterization of the output state and relation to experimentally accessible quantities is most conveniently achieved by the normally ordered electric field

correlations in the reduced coordinates τ_i ,

$$G_{n,m}^{\text{out}}(\tau_1, \dots, \tau_{n+m}) = \left\langle \mathcal{E} \left| \prod_{i=1}^n \psi^\dagger(\tau_i) \prod_{j=n+1}^{n+m} \psi(\tau_j) \right| \mathcal{E} \right\rangle.$$

These correlation functions are experimentally accessible in a homodyne detection scheme. The full expression for the correlations of the outgoing fields for an incoming coherent state is presented in the supplement. In the following, we provide the result for the two point correlation function $G_{0,2}^{\text{out}}$, which reduces to

$$G_{0,2}^{\text{out}}(\tau, \tau') = \mathcal{E}(\tau)\mathcal{E}(\tau') \exp[-i\varphi(\tau - \tau')] \quad (12)$$

$$\times \exp\left(\int du |\mathcal{E}(u)|^2 \left[e^{-i\varphi(u-\tau)-i\varphi(u-\tau')} - 1\right]\right).$$

We can distinguish two different contribution: first, we find a strong spatial correlation determined by the phase contribution $\varphi(\tau - \tau')$, which provides direct information about the effective interaction potential between the polaritons. It is this contribution, which allows the access to the effective interaction potential within a homodyne detection scheme. The last factor describes additional phase shift and the suppression due to quantum fluctuations, which are small corrections for $\xi_{\text{out}}|\mathcal{E}(\tau)|^2 \ll 1$.

A full characterization of the outgoing field for an incoming field coherent field \mathcal{E} is provided by the Wigner function $W(q, p)$. In contrast to circuit and cavity QED experiments, where the photons within the resonator are characterized by a single photonic mode [30, 31], our system here corresponds to a multimode setup. Therefore, in terms of the Wigner function, we can only express the reduced density matrix in a specific photonic mode. For this purpose, we define the annihilation operator for an arbitrary spatial mode $u(x)$ as $\hat{a}_u = \int dx u(x)\psi(x)$ and the related quadrature operators as $\hat{q} = (\hat{a}_u + \hat{a}_u^\dagger)/2$, $\hat{p} = (\hat{a}_u - \hat{a}_u^\dagger)/2i$. Then, the Wigner function derives directly from the analytical expression for the correlation functions $G_{n,m}^{\text{out}}$ for the incoming coherent field [32],

$$W(q, p) = \frac{2}{\pi} \sum_{nm} \frac{(-1)^{n+m}}{n!m!} \mathcal{G}_{nm} \partial_{\alpha^*}^n \partial_{\alpha}^m e^{-2|\alpha|^2} \quad (13)$$

with $\alpha = q + ip$, and \mathcal{G}_{nm} the overlap of the electric field correlations with the probe photonic mode

$$\mathcal{G}_{nm} = \int d^{n+m}\tau G_{n,m}^{\text{out}}(\tau_1, \dots, \tau_{n+m}) \prod_{i=1}^n u(\tau_i)^* \prod_{j=n+1}^{n+m} u(\tau_j).$$

In order to characterize short range correlations between photons we consider a homodyne detection [33–35] with $u(x)$ being a localized mode having size l_{probe} much shorter than ξ_{out} . The quasi-probability $W(q, p)$ for different strengths of the interaction is shown in Fig. 3. For weak interactions $\varphi(0) \ll 1$, the leading correction due to quantum fluctuations to the Gaussian coherent state is a small squeezing. However, for increasing interaction we

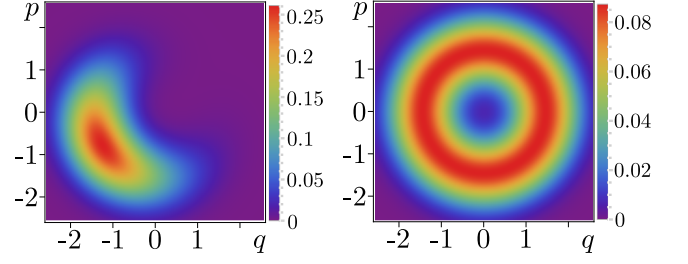


FIG. 3. Wigner function describing short range correlations ($l_{\text{probe}} \ll \xi_{\text{out}}$) for long-photons $l_{\text{coh}}/l_{\text{probe}} = 100$ for two different strengths of interaction: $\varphi(0) = \pi/64$ (left) and $\varphi(0) = \pi$ (right).

obtain a strongly mixed state. This behavior is a result of the localized measurement tracing out all positions outside the $u(x)$. Such an operation, acting on our strongly spatially entangled state, leads to the mixed state.

A crucial property of our analysis is that it demonstrates the possibility to probe the microscopic interaction potential between the Rydberg polaritons via a homodyne detection scheme for a coherent input state. This method can easily be extended to probe higher body interactions between the polaritons, which are expected to appear for higher polariton densities. Such a n -body interaction on the microscopic level takes the form

$$H_n = \frac{1}{n!} \int d\mathbf{x} U_n(x_1, \dots, x_n) \prod_{i=1}^n n(x_i) \psi^\dagger(x_i) \psi(x_i) \quad (14)$$

with the n -body interaction potential U_n . This term can be straightforwardly included in the exact solution. As an example, we present the results for a three-body interaction, which leads, in analogy to Eq. (7), to a phase contribution to the three photon wave-function

$$\phi^{\text{out}}(\tau_1, \tau_2, \tau_3) = e^{-i\varphi_3(\tau_1-\tau_2, \tau_2-\tau_3)} \phi^{\text{in}}(\tau_1, \tau_2, \tau_3).$$

The phase factor $\varphi_3(u, v)$ induced by the three-body interaction takes the form

$$\frac{1}{\hbar c} \int_{-\infty}^{\infty} dw \tilde{n}(w+u) \tilde{n}(w+v) \tilde{n}(w) \tilde{U}_3(w+u, w+v, w),$$

with \tilde{U}_3 defined in analogy to \tilde{V} . The corresponding three-body interaction potential can then be experimentally observed in a homodyne detection of the correlations $G_{0,3}^{\text{out}}$.

In conclusion, we studied Rydberg slow light polaritons and obtained a consistent quantum theory for a Kerr nonlinearity. For weak interactions we demonstrated that the system reduces to a conventional Kerr nonlinearity, while for moderate interactions we derived quantum corrections. Rydberg slow light polaritons naturally lead to a finite interaction range and a mass term, which regularize previous problems in deriving a quantum theory of a Kerr nonlinearity. Our approach provides a promising tool for the direct experimental observation of the

two-body interaction potential as well as higher body interaction potentials via a homodyne detection.

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- [1] X. Hu, P. Jiang, C. Ding, H. Yang., Q. Gong, H. Yang and Q. Gong, *Picosecond and low-power all-optical switching based on an organic photonic-bandgap microcavity*, Nat. Photon., **2**, 185–189 (2008).
 - [2] D. A. B. Miller, *Are optical transistors the logical next step?*, Nat. Photon., **4**, 3–5 (2010).
 - [3] G. J. Milburn, *Quantum optical Fredkin gate*, Phys. Rev. Lett., **62**, 2124–2127 (1989).
 - [4] H. J. Kimble, *The quantum internet.*, Nature, **453**, 1023–30 (2008).
 - [5] A. Muthukrishnan, M. O. Scully and M. S. Zubairy, *Quantum microscopy using photon correlations*, J. Opt. B, **6**, S575 (2004).
 - [6] V. Giovannetti, S. Lloyd and L. Maccone, *Advances in quantum metrology*, Nat. Photon., **5**, 222–229 (2011).
 - [7] R. W. Boyd, *Nonlinear Optics*, volume 5, Academic Press (2003).
 - [8] N. Matsuda, R. Shimizu, Y. Mitsumori, H. Kosaka and K. Edamatsu, *Observation of optical-fibre Kerr nonlinearity at the single-photon level*, Nat. Photonics, **3**, 95–98 (2009).
 - [9] Y. O. Dudin and A. Kuzmich, *Strongly interacting Rydberg excitations of a cold atomic gas.*, Science, **336**, 887–9 (2012).
 - [10] L. Li, Y. O. Dudin and A. Kuzmich, *Entanglement between light and an optical atomic excitation*, Nature, **498**, 466–469 (2013).
 - [11] S. Baur, D. Tiarks, G. Rempe, S. Dürr and D. Stephan, *Single-Photon Switch based on Rydberg Blockade*, 2013, **112**, 1–10 (2014).
 - [12] H. Gorniaczyk, C. Tresp, J. Schmidt, H. Fedder and S. Hofferberth, *Single Photon Transistor Mediated by Inter-State Rydberg Interaction*, Phys. Rev. Lett., **113**, 053601 (2014).
 - [13] D. Tiarks, S. Baur, K. Schneider, S. Dürr and G. Rempe, *Single-Photon Transistor Using a Förster Resonance*, Phys. Rev. Lett., **113**, 053602 (2014).
 - [14] H. Gorniaczyk, C. Tresp, P. Bienias, A. Paris-Mandoki, W. Li, I. Mirgorodskiy, H. P. Büchler, I. Lesanovsky and S. Hofferberth, *Enhancement of single-photon transistor by Stark-tuned Förster resonances*, ArXiv:1511.09445 (2015).
 - [15] T. Peyronel, O. Firstenberg, Q.-Y. Liang, S. Hofferberth, A. V. Gorshkov, T. Pohl, M. D. Lukin and V. Vuletić, *Quantum nonlinear optics with single photons enabled by strongly interacting atoms.*, Nature, **488**, 57–60 (2012).
 - [16] O. Firstenberg, T. Peyronel, Q.-y. Liang, A. V. Gorshkov, M. D. Lukin and V. Vuletić, *Attractive photons in a quantum nonlinear medium*, Nature, **502**, 71–75 (2013).
 - [17] J. Otterbach, M. Moos, D. Muth and M. Fleischhauer, *Wigner Crystallization of Single Photons in Cold Rydberg Ensembles*, Phys. Rev. Lett., **111** (2013).
 - [18] P. Bienias, S. Choi, O. Firstenberg, M. F. Maghrebi, M. Gullans, M. D. Lukin, A. V. Gorshkov and H. P. Büchler, *Scattering resonances and bound states for strongly interacting Rydberg polaritons*, Phys. Rev. A, **90**, 053804 (2014).
 - [19] A. V. Gorshkov, J. Otterbach, M. Fleischhauer, T. Pohl and M. D. Lukin, *Photon-Photon Interactions via Rydberg Blockade*, Phys. Rev. Lett., **107**, 133602 (2011).
 - [20] B. He, A. V. Sharypov, J. Sheng, C. Simon and M. Xiao, *Two-Photon Dynamics in Coherent Rydberg Atomic Ensemble*, Phys. Rev. Lett., **112**, 133606 (2014).
 - [21] M. Moos, M. Höning, R. Unanyan and M. Fleischhauer, *Many-body physics of Rydberg dark-state polaritons in the strongly interacting regime*, Phys. Rev. A, **92**, 053846 (2015).
 - [22] M. F. Maghrebi, N. Y. Yao, M. Hafezi, T. Pohl, O. Firstenberg and A. V. Gorshkov, *Fractional quantum Hall states of Rydberg polaritons*, Phys. Rev. A, **91**, 033838 (2015).
 - [23] M. F. Maghrebi, M. J. Gullans, P. Bienias, S. Choi, I. Martin, O. Firstenberg, M. D. Lukin, H. P. Büchler and A. V. Gorshkov, *Coulomb bound states of strongly interacting photons*, Phys. Rev. Lett., **115**, 123601 (2015).
 - [24] K. J. Blow, R. Loudon and S. J. D. Phoenix, *Exact solution for quantum self-phase modulation*, J. Opt. Soc. Am. B, **8**, 1750 (1991).
 - [25] F. X. Kartner, L. Joneckis and H. A. Haus, *Classical and quantum dynamics of a pulse in a dispersionless nonlinear fibre*, Quantum Opt. J. Eur. Opt. Soc. Part B, **4**, 379–396 (1992).
 - [26] L. G. Joneckis and J. H. Shapiro, *Quantum propagation in a Kerr medium: lossless, dispersionless fiber*, J. Opt. Soc. Am. B, **10**, 1102 (1993).
 - [27] L. Boivin, F. Kärtner and H. Haus, *Analytical Solution to the Quantum Field Theory of Self-Phase Modulation with a Finite Response Time*, Phys. Rev. Lett., **73**, 240–243 (1994).
 - [28] J. Shapiro, *Single-photon Kerr nonlinearities do not help quantum computation*, Phys. Rev. A, **73**, 062305 (2006).
 - [29] K. Jachymski, P. Bienias and H. P. Büchler, *Three-body interaction of Rydberg slow light polaritons*, arXiv:1604.03743 (2016).
 - [30] A. Wallraff, D. I. Schuster, A. Blais, L. Frunzio, R.-S. Huang, J. Majer, S. Kumar, S. M. Girvin and R. J. Schoelkopf, *Strong coupling of a single photon to a superconducting qubit using circuit quantum electrodynamics.*, Nature, **431**, 162–167 (2004).
 - [31] S. Haroche and J.-M. Raimond, *Exploring the Quantum Atoms, Cavities, and Photons*, Oxford Univ. Press (2006).
 - [32] *See Supplementary Material for detailed derivation.*
 - [33] S. Deléglise, I. Dotsenko, C. Sayrin, J. Bernu, M. Brune, J.-M. Raimond and S. Haroche, *Reconstruction of non-classical cavity field states with snapshots of their decoherence.*, Nature, **455**, 510–4 (2008).
 - [34] A. I. Lvovsky and M. G. Raymer, *Continuous-variable optical quantum-state tomography*, Rev. Mod. Phys., **81**, 299–332 (2009).
 - [35] E. Bimbard, R. Boddada, N. Vitrant, A. Grankin, V. Parigi, J. Stanojevic, A. Ourjoumtsev and P. Grangier, *Homodyne Tomography of a Single Photon Retrieved on Demand from a Cavity-Enhanced Cold Atom Memory*, Phys. Rev. Lett., **112**, 033601 (2014).

- [36] D. F. Walls and G. J. Milburn, *Quantum Optics*, Springer-Verlag (2008).

SUPPLEMENTAL MATERIAL

Regime of parameters in which mass term is negligible

In this section, we derive a regime of parameters in which we can drop the mass term in the polaritonic dispersion relation. For this purpose, we analyze two polaritons propagating through a cloud of atoms with a constant density. In the relative r and center of mass R coordinates, the Schrödinger equation for the two-body wave function $\phi(R, r)$ takes the form [18]

$$\hbar\omega\phi(R, r) = \left(-i\hbar v_g \partial_R - \frac{\hbar^2}{m} \partial_r^2 + \alpha V(r)\right) \phi(R, r), \quad (\text{S1})$$

where

$$m = \hbar \frac{(g^2 + \Omega^2)^3}{2c^2 g^2 \Delta \Omega^2}, \quad V(r) = \frac{2\Omega^2 \hbar}{\delta} \frac{1}{1 + r^6/\xi^6}, \quad \alpha = \frac{g^4}{(g^2 + \Omega^2)^2}, \quad v_g = \frac{\Omega^2}{\Omega^2 + g^2} c. \quad (\text{S2})$$

Assuming that the mass term is negligible, we can find the analytical solution of Eq. S1 of the form

$$\phi_0(R, r) = \phi(0, r) \exp\left(i \frac{\omega}{v_g} R - i\varphi(0) \frac{R}{L} \frac{1}{1 + (r/\xi)^6}\right) \quad (\text{S3})$$

where we used the relation $\varphi(0) = L\alpha V(0)/v_g$, see Eq. 9. For latter purposes let us define $\varphi_{\text{int}}(R, r) = \varphi(0)R/L/(1 + (r/\xi)^6)$. Using the above solution, we calculate perturbatively the corrections due to the mass term to the phase shift. For this purpose we express the full solution ϕ using ϕ_0 :

$$\phi(R, r) = \phi_0(R, r) e^{-i\vartheta_m(R, r)}, \quad (\text{S4})$$

where $\vartheta_m(R, r)$ takes into account the impact of the mass term. Next, we insert this Ansatz into Eq. S1. Exploiting that ϕ_0 is the solution for the unperturbed Hamiltonian, most of the terms cancel and we arrive with the equation for ϑ_m :

$$0 = -iv_g \phi_0(R, r) \partial_R e^{-i\vartheta_m(R, r)} - \frac{\hbar^2}{m} \partial_r^2 \phi_0(R, r) e^{-i\vartheta_m(R, r)}. \quad (\text{S5})$$

This equation can be simplified once we take into account that in the perturbative limit $|\partial_r \vartheta_m(R, r)| \ll |\partial_r \varphi_{\text{int}}(R, r)|$ and $|\partial_r^2 \vartheta_m(R, r)| \ll |\partial_r^2 \varphi_{\text{int}}(R, r)|$. Moreover, considered photons are much longer than ξ_{out} and, therefore, we drop $\partial_r \phi(0, r)$ and $\partial_r^2 \phi(0, r)$ terms. Finally, Eq. S5 simplifies to

$$0 = -iv_g \phi_0(R, r) \partial_R e^{-i\vartheta_m(R, r)} - \frac{\hbar^2}{m} \phi(0, r) e^{-i\vartheta_m(R, r)} e^{i \frac{\omega}{v_g} R} \partial_r^2 \exp\left(-i\varphi(0) \frac{R}{L} \frac{1}{1 + (r/\xi)^6}\right). \quad (\text{S6})$$

This equation leads to the solution for $\vartheta_m(L, r)$ of the form

$$\vartheta_m(L, r) = -\frac{1}{v_g} \int_0^L dR \frac{\hbar^2 \partial_r^2 \exp\left(-i\varphi(0) \frac{R}{L} \frac{1}{1 + (r/\xi)^6}\right)}{\exp\left(-i\varphi(0) \frac{R}{L} \frac{1}{1 + (r/\xi)^6}\right)}. \quad (\text{S7})$$

In order to estimate $\vartheta_m(R = L, r)$ we consider its value at $r = \xi$, which is equal to

$$\vartheta_m(L, \xi) = 3(\varphi(0) + i) \frac{L^2}{\xi^2} \frac{g^6}{(g^2 + \Omega^2)^3}, \quad (\text{S8})$$

and corresponds to the result for φ_m from the main text. We see that the mass term can be dropped if two conditions $\varphi_m \ll 1$ and $\varphi_m/\varphi(0) \ll 1$ are satisfied.

Correlations of the outgoing fields for an incoming coherent state

Here, we derive the general expression for the correlations $G_{n,m}^{\text{out}}$ of the outgoing fields for an incoming coherent state $|\mathcal{E}\rangle$. We start by inserting the exact solution for bosonic field operators $\hat{\psi}(z, t) = e^{-i\hat{J}(z,t)}\hat{\psi}_0(z - ct)$ into the definition of $G_{n,m}^{\text{out}}$ from the main text. This leads to

$$G_{n,m}^{\text{out}}(\tau_1, \dots, \tau_{n+m}) = \left\langle \mathcal{E} \left| \prod_{i=1}^n \left(e^{-i\hat{J}(z_i,t)} \hat{\psi}_0(z_i - ct) \right)^\dagger \prod_{j=n+1}^{n+m} e^{-i\hat{J}(z_j,t)} \hat{\psi}_0(z_j - ct) \right| \mathcal{E} \right\rangle, \quad (\text{S9})$$

where $\tau_i = z_i - ct$. Our goal is to transform the product of operators to the normally ordered expression, of which expectation value in a coherent state is trivial. For this purpose, we first use the relation

$$\hat{\psi}_0(z_i - ct) e^{-i\hat{J}(z_j,t)} = e^{-i\hat{J}(z_j,t)} e^{-i\varphi(z_i - z_j)} \hat{\psi}_0(z_i - ct) \quad (\text{S10})$$

to normally order the ψ_0 operators in the Eq. S9,

$$\begin{aligned} G_{n,m}^{\text{out}}(\tau_1, \dots, \tau_{n+m}) &= \left\langle \mathcal{E} \left| \prod_{k=1}^n \hat{\psi}_0^\dagger(\tau_k) \prod_{i=1}^n e^{i\hat{J}(z_i,t)} \prod_{j=n+1}^{n+m} e^{-i\hat{J}(z_j,t)} \prod_{l=n+1}^{n+m} \hat{\psi}_0(\tau_l) \right| \mathcal{E} \right\rangle \\ &\times \exp \left[i \sum_{k>l=1}^n \varphi(\tau_k - \tau_l) \right] \exp \left[-i \sum_{k>l=n+1}^{m+n} \varphi(\tau_k - \tau_l) \right]. \end{aligned} \quad (\text{S11})$$

Next, we use the fact that in the limit of $t \rightarrow \infty$ the expression for $\hat{J}(z_i, t)$ can be written as $\hat{J}(z_i, t) = \int_{-\infty}^{\infty} du \hat{I}(u) \varphi(u - z_i + ct)$, and that $\hat{J}(z_j, t)$ commutes with $\hat{J}(z_i, t)$, in order to rewrite the product of exponentials appearing in Eq. S11 in the following way:

$$\begin{aligned} \prod_{i=1}^n e^{i\hat{J}(z_i,t)} \prod_{j=n+1}^{n+m} e^{-i\hat{J}(z_j,t)} &= \exp \left(i \sum_{i=1}^n \hat{J}(z_i, t) - i \sum_{j=n+1}^{n+m} \hat{J}(z_j, t) \right) \\ &= \exp \left(\int_{-\infty}^{\infty} du \hat{I}(u) \left[i \sum_{i=1}^n \varphi(u - \tau_i) - i \sum_{j=n+1}^{n+m} \varphi(u - \tau_j) \right] \right). \end{aligned} \quad (\text{S12})$$

The last expression can be transformed to the normally ordered one using the relation [27]:

$$\exp \left(\int du g(u) \hat{I}(u) \right) =: \exp \left(\int du \left(e^{g(u)} - 1 \right) \hat{I}(u) \right) :. \quad (\text{S13})$$

In our case $g(u) = i \sum_{i=1}^n \varphi(u - \tau_i) - i \sum_{j=n+1}^{n+m} \varphi(u - \tau_j)$, what leads to

$$\prod_{i=1}^n e^{i\hat{J}(z_i,t)} \prod_{j=n+1}^{n+m} e^{-i\hat{J}(z_j,t)} =: \exp \left(\int_{-\infty}^{\infty} du \hat{I}(u) \left[\exp \left(i \sum_{i=1}^n \varphi(u - \tau_i) - i \sum_{j=n+1}^{n+m} \varphi(u - \tau_j) \right) - 1 \right] \right) :. \quad (\text{S14})$$

The last equation inserted into the Eq. S11 provides the final result,

$$\begin{aligned} G_{n,m}^{\text{out}}(\tau_1, \dots, \tau_{n+m}) &= \prod_{i=1}^n \hat{\mathcal{E}}^*(\tau_i) \prod_{j=n+1}^{n+m} \hat{\mathcal{E}}(\tau_j) \exp \left[i \sum_{k>l=1}^n \varphi(\tau_k - \tau_l) \right] \exp \left[-i \sum_{k>l=n+1}^{m+n} \varphi(\tau_k - \tau_l) \right] \\ &\times \exp \left(\int_{-\infty}^{\infty} du |\mathcal{E}(u)|^2 \left[\exp \left(i \sum_{i=1}^n \varphi(u - \tau_i) - i \sum_{j=n+1}^{n+m} \varphi(u - \tau_j) \right) - 1 \right] \right). \end{aligned} \quad (\text{S15})$$

Two special cases of these correlations, i.e., $G_{0,1}^{\text{out}}$ and $G_{0,2}^{\text{out}}$ are presented in the main text, see Eqs 10 and 12, respectively.

Wigner function from correlation functions

Here, we show how Wigner function $W(q, p)$ can be calculate using the correlation functions \mathcal{G}_{nm} . Our starting point is symmetrically ordered characteristic function $\chi(\eta)$ defined as

$$\chi(\eta) = \text{Tr} [\rho \exp [\eta \hat{a}_u^\dagger - \eta^* \hat{a}_u]] . \quad (\text{S16})$$

The function $\chi(n)$ can be expressed using correlation function $\mathcal{G}_{nm} = \langle (\hat{a}_u^\dagger)^n \hat{a}_u^m \rangle$ as

$$\chi(\eta) = \sum_{nm} \frac{\eta^n (-\eta^*)^m}{n!m!} e^{-|\eta|^2/2} \text{Tr} [\rho (\hat{a}_u^\dagger)^n \hat{a}_u^m] = \sum_{nm} \frac{\eta^n (-\eta^*)^m}{n!m!} e^{-|\eta|^2/2} \mathcal{G}_{nm}. \quad (\text{S17})$$

The Wigner function is defined as the Fourier transform of the characteristic function $\chi(\eta)$ [36],

$$W(\alpha) = \frac{1}{\pi^2} \int d^2\eta e^{\eta^* \alpha - \eta \alpha^*} \chi(\eta). \quad (\text{S18})$$

Finally, we insert $\chi(\eta)$ from Eq. S17 into the definition S18 and afterwards transform $W(\alpha)$ to a more concise expression:

$$\begin{aligned} W(\alpha) &= \frac{1}{\pi^2} \sum_{nm} \frac{(-1)^m}{n!m!} \int d^2\eta e^{\eta^* \alpha - \eta \alpha^*} \eta^n (\eta^*)^m e^{-|\eta|^2/2} \mathcal{G}_{nm} \\ &= \frac{1}{\pi^2} \sum_{nm} \frac{(-1)^{n+m}}{n!m!} \mathcal{G}_{nm} \partial_{\alpha^*}^n \partial_{\alpha}^m \int d^2\eta e^{\eta^* \alpha - \eta \alpha^*} e^{-|\eta|^2/2} \\ &= \frac{2}{\pi} \sum_{nm} \frac{(-1)^{n+m}}{n!m!} \mathcal{G}_{nm} \partial_{\alpha^*}^n \partial_{\alpha}^m e^{-2|\alpha|^2}, \end{aligned} \quad (\text{S19})$$

which is the formula presented in the main text.